*FUNDAMENTAL PROBLEMS IN STATISTICAL MECHANICS VII*
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# Computer Simulations in Statistical Physics [1]

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## 1 INTRODUCTION

The aim of these lectures is *not* to give a review of computer simulations in statistical physics. The number of applications of computer simulations has grown tremendously over the past two decades to the extent that simulations now probably provide more 'experimental' data to test statistical mechanical theories than real experiments. For this reason the present lectures focus on applications of computer simulations that illustrate the different ways in which computer simulations can be used to gain a better understanding of the complex many-body systems that are studied in statistical physics.

The first question that should be addressed in these lectures is : *Why do computer simulations at all?*. This question is important because the numerical studies that we perform in the context of statistical mechanics are not of the same nature as in, say, material science. In particular, it is usually not our aim to simulate a particular material as accurately as possible. Rather, we tend to study simplified models that contain the essential features of the many-body systems that we wish to understand. We use computer simulations mainly for two reasons:

1. To test theoretical approximations

2. To 'discover' new phenomena, i.e. to gather the building blocks for new theories.

However, computer simulations themselves are no substitute for a theoretical description. On the contrary, the ultimate aim of any simulation should be to make itself redundant: if a simple theory can be constructed that accounts for the computer simulation results, then clearly such a theory is preferable to a computer program.

In these lectures we shall mainly consider the first application of computer simulation. In particular, we shall discuss some recent numerical techniques that enable us to test theories of transport phenomena in simple fluids in great detail. An example of the other application of computer simulations in the exploration of novel phenomena is discussed in considerable detail in a recent review article on computer simulations of liquid crystals [1].

## 2 LONG-TIME TAILS

### 2.1 Introduction

In the history of the kinetic theory of fluids, 1969-1970 was a crucial year. In that year Alder and Wainwright [2] published a paper in which they demonstrated the breakdown of the 'Molecular Chaos' assumption. The Molecular Chaos assumption, originally introduced

[1] This paper is based on material that has either been published elsewhere or has been submitted for publication.

by Boltzmann as the 'Stoßzahlansatz', states that the collisions experienced by a molecule in a fluid are uncorrelated. One consequence of this assumption is that the velocity autocorrelation function (VACF) of a tagged particle in fluid should decay exponentially. What Alder and Wainwright found is that the VACF of a particle in a moderately dense fluid of hard spheres or hard disks does not decay exponentially but algebraically. These algebraic long-time tails are the consequence of coupling between particle diffusion and shear modes in the fluid.

The Alder-Wainwright simulations caused a complete overhaul of the kinetic theory of dense fluids. The subsequent theoretical analyses of algebraic long-time tails were either based on an extension of kinetic theory [3] or on mode-coupling theory [4]. For a review, see ref. [5] . In the mode-coupling theory by Ernst, Hauge and van Leeuwen [4], it is assumed that the long-time tail is the consequence of coupling between particle diffusion and shear modes in the fluid. To a first approximation the leading term in the long-time tail of the velocity ACF is given by:

$$< v_x(0)v_x(t) > \approx \frac{D-1}{D} \frac{1}{\rho(4\pi(D_0+\nu_0)t)^{D/2}} \equiv \frac{d_0}{t^{D/2}}\,, \tag{1}$$

where $\rho$ is the number density, $D_0$ the 'bare' self-diffusion constant, $\nu_0$ the kinematic viscosity and $D$ the dimensionality.

Following this theoretical work, simulations were performed by Levesque and Ashurst [6] and, most extensively, by Erpenbeck and Wood [7,8] with the aim to verify the validity of eqn. 1. For three-dimensional fluids these simulations are extremely expensive because very long simulations on very large systems must be performed. Even so, the system sizes studied in the simulations of Erpenbeck and Wood were such that it was essential to apply finite-size corrections to the corresponding mode-coupling theory before a meaningful comparison with the simulations could be made. Following such an approach, Erpenbeck and Wood found agreement between their simulation results for the VACF and a finite-size mode-coupling theory for a number of different densities. Nevertheless the statistical accuracy of their data was such that it was not meaningful to verify either the value of the exponent of the algebraic tail or the functional form of the density-dependent tail coefficient independently.

In the case of two-dimensional fluids there is another problem. Ever since the discovery of hydrodynamic tails, it has been realized that a consistent description of mode-coupling effects in a two-dimensional fluid would result in a long-time tail that decays faster than $t^{-1}$, because in $2D$ the self-diffusion constant itself diverges. In fact, de Schepper and Ernst [9] computed the coefficient ($d_1$ in eqn. 2) of the first correction to the $t^{-1}$ tail for a system of hard disks. They predicted that this correction should be negative and proportional to $\log(t/t_0)/t$ (where $t_0^{-1}$ is the initial decay rate of the velocity ACF):

$$< v_x(0)v_x(t) > \approx d_0/t + d_1 \log(t/t_0)/t + \ldots \text{for } t/t_0 \gg 1. \tag{2}$$

However, this prediction is only expected to hold for times that are not too long. Forster et al. [10] argued that as $t \to \infty$, the tail should be renormalized to $1/(t\sqrt{\log t})$.

Thus far it has not been possible to compare these predictions directly with computer simulation data. The reason is that such a comparison requires accurate knowledge of the velocity ACF for very long times (many tens to hundreds of collision times). This requires very long simulations on very large systems (to avoid problems due to spurious correlations caused carried by sound waves in periodic systems). Thus far such calculations have remained beyond the power of presentday computers.

## 2.2 Lattice gases

In the remainder of this paper we shall indicate how computer simulations on lattice gases can be used to test theoretical predictions about 'long-time tails' in the velocity autocorrelation function of tagged particle in a fluid.

One of the attractive features of lattice gas models is that they are ideally suited to serve as a testing ground for concepts in kinetic theory. The reason is twofold: on the one hand the very simple structure of most lattice-gas models often makes it possible to work out in closed form the consequences of a particular approximation scheme in kinetic theory. On the other hand, lattice-gas models are ideally suited for computer simulation. Thus approximate schemes in kinetic theory can be tried out on lattice gas models before applying them to more realistic models of fluids or solids. Below we show how Lattice Gas Cellular Automata (LGCA's) can be used to verify in great detail the theoretical predictions about long-time tails in the the velocity autocorrelation function of tagged particles.

Lattice gas cellular automata have recently received a lot of attention because these model systems may provide a 'cheap' alternative to simulate the hydrodynamic behaviour of simple fluids [11]. Because of their simple structure which makes it comparatively easy to work out the consequences of a particular approximation scheme, LGCA's are, in principle, ideally suited to serve as a testing ground for concepts in kinetic theory. It is therefore only natural that lattice gas models were considered as promising systems to study the long-time behaviour of the tagged-particle VACF. This approach has been tried by Boon and Noullez [12] and Binder and d'Humières [13] for 2D systems. However, somewhat disappointingly, the statistical accuracy of these (long) numerical simulations is poor. Hence the presence of a long-time tail was hidden in the statistical noise and a special analysis was required to demonstrate that the simulation data are, in fact, compatible with the presence of an algebraic tail of approximately the expected amplitude [14].

However, using a technique that was recently developed by Frenkel [15,16] it is now possible to compute tagged particle VACF's in lattice gases with an efficiency that is a factor $10^6$ to $10^{10}$ higher than the earlier 'brute force' schemes. In the present paper we shall discuss the application of this new technique to the study of long-time tails.

In order to explain the numerical scheme, we first describe its application to lattice Lorentz gases. Thereafter we indicate how the method can be extended to study long-time tails in the velocity autocorrelation function of a tagged particle in a lattice gas cellular automaton.

## 2.3 Lattice Lorentz gases

Let us first consider a lattice-Lorentz gas with 'bond' disorder. This is a model system where a random walker performs a hopping motion along the nearest-neighbor bonds of a square (2D) or simple-cubic (3D) lattice. The moving particle can jump with equal probability along any of the 4 (or 6) bonds to a neighboring site. However, if the bond is 'broken', the particle remains at its original site until the next trial move. In the bond-disorder model, a finite fraction of the bonds is broken. The broken bonds are distributed randomly over the lattice. Two equivalent versions of this random walk process can be formulated: a) a discrete time version in which a new trial move is attempted every unit time and b) a continuous time version where the waiting times between successive trial moves are Poisson distributed. It is clear that statistical properties of the continuous-time

model, such as a time correlation function $C(t)$, can be obtained as a weighted average over all n-step correlations of the corresponding discrete-time model $C_D(n)$:

$$C(t) = \frac{1}{2}C_D(0)\delta(0_+) + \sum_{n=1}^{\infty} C_D(n)\frac{t^n}{n!}\exp(-t), \tag{3}$$

assuming an average rate of one jump per unit time for the continuous time model. It turns out that the theory for transport in lattice Lorentz gases is formulated most compactly for the continuous-time model, whereas the simulations are performed most conveniently for the discrete-time version.

Let us now consider the numerical computation of the velocity correlation function of a lattice-Lorentz gas. The discrete nature of the model leads to the following natural definitions of the 'velocity': the velocity $v_\alpha(t_n)$ of a random walker at discrete time $t_n$ in direction $\alpha$ is $+1(-1)$ if the random walker performed a jump in the $+\alpha(-\alpha)$ direction in time interval n, and is 0 otherwise. With this definition it is easy to see how to compute the velocity auto-correlation function $\phi(t) \equiv < \mathbf{v}(0).\mathbf{v}(t_n) >$. In principle this quantity can be computed by considering all possible starting positions of the random walker at time $t = 0$ and then averaging over all possible random walks that this particle can perform in a time interval $t_n$. The problem with this 'brute force' approach is that very many random walks must be sampled to obtain a reasonable statistical accuracy. To give a specific example: for a 2D lattice-Lorentz gas with a low concentration of broken bonds (say $p$=1%), the long time tail in the velocity ACF is expected to decay as $\phi(t) \approx p/t^2$. Hence, if we are interested in the long-time behaviour (e.g. $t = 100$), the amplitude of $\phi(t)$ will be $O(10^{-6})$. In order to obtain a signal-to-noise ratio of $O(1)$ we should therefore sample over $O(10^{12})$ independent random walks. Using an efficient random walk procedures [25] such a simulation would require some $10^2$ hours of CPU time on a Cyber 205 vector computer.

In fact, a method has been developed to reduce the required computer time by a factor $10^2$ [17] , but this technique will not work for lattice-gas cellular automata, to be considered below. Anyway, the approach described in the present paper results in far greater computational gains.

Let us consider the random walk problem that we wish to solve in some detail. In order to compute $< \mathbf{v}(0).\mathbf{v}(t) >$ we could pick a site $\mathbf{X}$ on the lattice at $t = 1/2$ (in order to minimize confusion we compute velocities at integer times and positions at half integer times) pick a point $\mathbf{X}'$ at time $t = t_n - \frac{1}{2}$ that can be reached in a possible random walk in time $t_n - 1$, compute the product $\mathbf{v}(0).\mathbf{v}(t_n)$ and average over: 1) all possible random walks linking $\mathbf{X}(t = \frac{1}{2})$ and $\mathbf{X}'(t = t_n - \frac{1}{2})$, 2) all $\mathbf{X}'$ and 3) all $\mathbf{X}$.

First, consider the computation of the velocity at $t_n$. If the random walker is at site $\mathbf{X}'$ at time $t_n - \frac{1}{2}$ then it can jump to any of the neighboring sites with equal probability, unless any of the bonds connected to $\mathbf{X}'$ is broken. If all bonds connected to $\mathbf{X}'$ are open, then clearly the average velocity of the random walker in the time interval centered at $t_n$ is 0. Only if one or more bonds are broken can the random walker have a non-zero average velocity in the interval $[t_n - \frac{1}{2}, t_n + \frac{1}{2}]$. Let us denote this velocity by $\mathbf{v}(\mathbf{X}')$. Using this definition, we note that the average velocity of a random walker at time $t = 0$ (i.e. just before its arrival at $\mathbf{X}$ is equal to $-\mathbf{v}(\mathbf{X})$. Clearly, if there are no broken bonds around $\mathbf{X}$, $\mathbf{v}(\mathbf{X}) = 0$ and all random walks starting at $\mathbf{X}$ do not contribute to $< \mathbf{v}(0).\mathbf{v}(t) >$. From now on we restrict our attention to those random walks for which $\mathbf{v}(\mathbf{X}) \neq 0$. The actual number of lattice sites for which $\mathbf{v}(\mathbf{X}) \neq 0$ can easily be counted once the distribution of broken bonds has been specified. The average fraction of sites for which $\mathbf{v}_\alpha(\mathbf{X}) \neq 0$. for any direction $\alpha$ is equal to $2p(1 - p)$, where $p$ is the fraction of broken bonds.

Now let us consider the computation of $< \mathbf{v}(0).\mathbf{v}(t) >$. For $t = 0$ the answer is simply $(1-p)$. For $t = 1$, we obtain $-\overline{v^2(X)}$ where the bar denotes averaging over all lattice sites. In order to compute $< \mathbf{v}(0).\mathbf{v}(t) >$ for $t > 1$, we note that if at time $t - \frac{1}{2}$ a particle is at position $\mathbf{X}$ then after one more time-step, it has a probability $1/2D$ ($D$ = dimensionality) to be on any of the $\ell$ ($\leq 2D$) connected nearest neighbor sites of $\mathbf{X}$ and a probability $(2D-\ell)/2D$ to be still at site $\mathbf{X}$. The average velocity of this random walker at time $t+1$ is therefore :

$$\mathbf{v}(t+1) = \sum_{\text{all } \mathbf{X}' \text{ connected to } \mathbf{X}} \frac{1}{2D}\mathbf{v}(\mathbf{X}') \tag{4}$$

where the original site $\mathbf{X}$ is counted as 'connected' to itself for every broken bond present. We now attribute this average velocity $\mathbf{v}(t+1)$ to the site $\mathbf{X}$ and denote it by $\mathbf{v^{(2)}}(\mathbf{X})$. Clearly, $< \mathbf{v}(0).\mathbf{v}(2) >= -\overline{\mathbf{v}(\mathbf{X}).\mathbf{v^{(2)}}(\mathbf{X})}$. Analogously, by iteration we obtain $\mathbf{v^{(3)}}(\mathbf{X})$ from $\mathbf{v^{(2)}}(\mathbf{X})$, and so on.

Note that in this way we compute the contribution to $< \mathbf{v}(0).\mathbf{v}(t) >$ from *all* possible random walks. Hence, once we have specified the distribution of broken bonds on the lattice, the present computation of $< \mathbf{v}(0).\mathbf{v}(t) >$ is *exact*. Statistical errors are only due to the fact that the velocity correlation function should be computed for a large number of independently generated realizations of the bond disorder. The other point to note is that the iteration scheme requires only very simple matrix operations that are eminently vectorizable. To demonstrate the power of the present method, figure 1 shows the long-time tail of the velocity ACF of a $2D$ lattice-Lorentz gas with bond disorder.

Note that the statistical error (points) in figure 1 decreases with increasing time. For the longest times studied in this simulation the statistical error is less than 1 in $10^{10}$. The calculation required 13 minutes of CPU-time on a CYBER 205 vector computer. To achieve the same accuracy by straightforward Monte Carlo sampling would require over $10^5$ years of CPU time on the same machine. The difference between the statistical error obtained by the present method and those obtained using the Monte Carlo approach [17] is particularly striking for long times. Direct sampling over trajectories yields a statistical error that is essentially independent of time. Hence to compute a $t^{-2}$ long-time tail over a time interval that is 10 times longer required a factor $10^4$ (!) more computing time ($10^5$ in 3D). In contrast, the error in the present matrix-method decreases with time. Hence, to study a 10-times longer correlation interval simple requires a run that is 10 times longer. The relative merit of the present method becomes greater at longer times. In our case the gain with respect to the random walk method was a factor $10^{10}$, but the comparison could have been made even more biased by going to longer times.

## 2.4 Lattice Gas Cellular Automata

Next we shall show how, by a slight extension, the present method can be applied to compute the long-time behaviour of velocity ACF's in lattice-gas cellular automata (LGCA's).

At first sight, the computation of the velocity ACF for a 'particle' in a system of lattice-gas CA's seems very different from the corresponding problem of a random walker in a lattice- Lorentz gas. After all, lattice gas cellular automata are meant to model systems of moving, interacting particles. In particular, LGCA's have been constructed as a highly simplified model for an atomic fluid. In the present paper we shall focus on the properties of LGCA's on a $2D$ triangular lattice (FHP-model [11,21] and on a $3D$ face-centered hyper-cubic (FCHC) lattice.

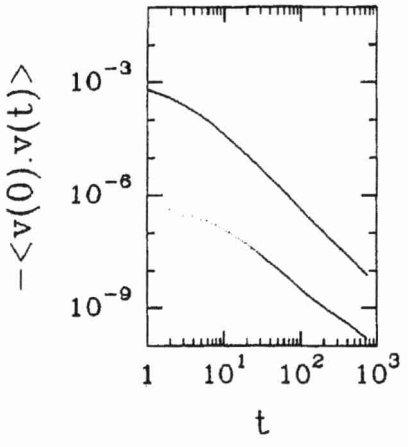


Figure 1: Velocity autocorrelation function of a random walker on a two-dimensional square lattice-Lorentz gas with a fraction of 1% broken bonds. The drawn curve is the velocity correlation function. The dots represent the statistical error.

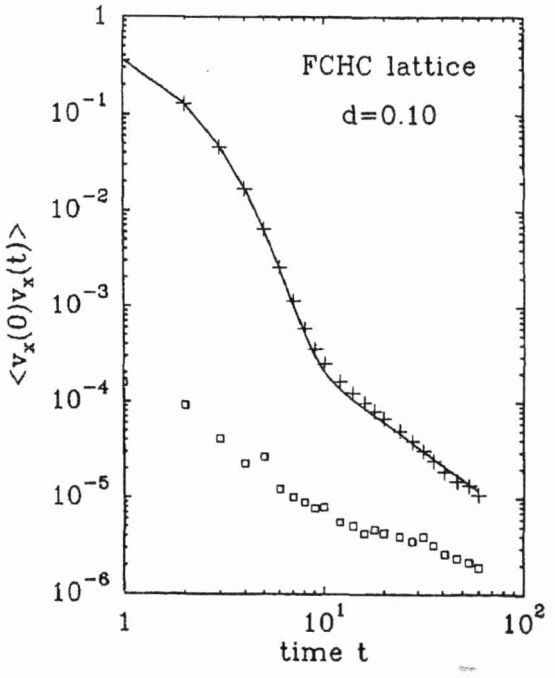


Figure 2: Log-log plot of the normalized velocity autocorrelation function of a 3-dimensional lattice gas cellular automaton on a FCHC lattice for density $d = 0.10$. The density $d$ is in units of average number of particles per link. The time is in units of the time between successive updates of the lattice gas cellular automaton $\Delta t(=1)$. The solid line is the prediction of mode-coupling theory, the crosses are the simulation data. Note that the estimated error (open squares) decreases with increasing $t$ to a value of order $10^{-6}$.

In LGCA's, the particles are constrained to move along the bonds joining the lattice sites. No two particles can move along the same bond in the same direction. The state of the lattice is completely specified by indicating which links are occupied and which are empty. This implies that lattice-gas particles are indistinguishable.

The time evolution of the system is governed by the following rules:

1. Propagation: all particles move in one time step (for convenience we choose $\Delta t = 1$) from their initial lattice position (say $\mathbf{X}$) to a new position ($\mathbf{X}' = \mathbf{X} + \mathbf{c}_\alpha$; where $\mathbf{c}_\alpha$ is the velocity of species $\alpha$). The velocities $\mathbf{c}_\alpha$ are such that at the end of the propagation steps all particles are once more positioned at lattice sites.

2. Collision: the particles at all sites on the lattice undergo a collision that converves the total number of particles and the total momentum at each site. Usually, these local collision rules are deterministic.

Provided that the lattice has a sufficiently high symmetry (e.g. triangular in 2 dimensions) and the collision rules are sufficiently isotropic (for a discussion, see [18]), it can be shown that the equation that governs the time evolution of the distribution function of such a lattice gas becomes equivalent to the Navier-Stokes equation for an incompressible fluid in the limit that the flow velocity is much less than the particle velocity, and all spatial variations in the system occur on a scale that is large compared to the mean free path of the lattice gas particles. In this respect LGCA's model atomic fluids.

When attempting to compute the velocity correlation function of a particle in a lattice gas CA, one is immediately confronted with a conceptual problem. As all lattice gas particles are indistinguishable, the velocity correlation function of 'a particle' is ill defined. As soon as a particle has collided it is no longer possible to identify any of the outgoing particles as the original particles whose velocity ACF we are attempting to compute. To avoid this problem, the particle under consideration must be labeled differently from the rest (say, a 'blue' particle in a sea of 'red' particles). Once the collision rules for all particles have been specified we can then compute the velocity ACF of a single tagged particle. This is the approach that has been pursued by Boon and Noullez for the FHP model [12] and by Binder and d'Humières for the HPP model [13]. As mentioned above, this approach yields poor statistics because we must solve the dynamics of all N lattice-gas particles in order to follow the time-evolution of 1 tagged particle.

An alternative approach that effectively side-steps the problem referred to above has been followed by Colvin, Ladd and Alder [26]. These authors compute not the velocity ACF of a tagged particle but the autocorrelation function of the fluid velocity at a lattice site. This method yields somewhat better statistics and made it possible to observe a long-time tail in the site-velocity ACF. However, the site-velocity ACF does not contain the same mode-coupling contributions as the tagged-particle velocity ACF and is therefore, from a theoretical point of view, of less interest.

Fortunately, the matrix method described in the previous section can be extended in such a way that it becomes possible the compute the velocity ACF of a tagged particle in a lattice-gas cellular automaton with high accuracy. In order to do so we have to impose one restriction on the rules of the lattice gas automaton, namely that the collision rules for a tagged particle with untagged particles result in the occupation of the same output states as in the case of collisions between untagged particles. And, most importantly, the tagged particle has equal probability to be in any of the occupied output states. Hence, for the tagged particle the collision rules are stochastic, although for a 'colour-blind' observer,

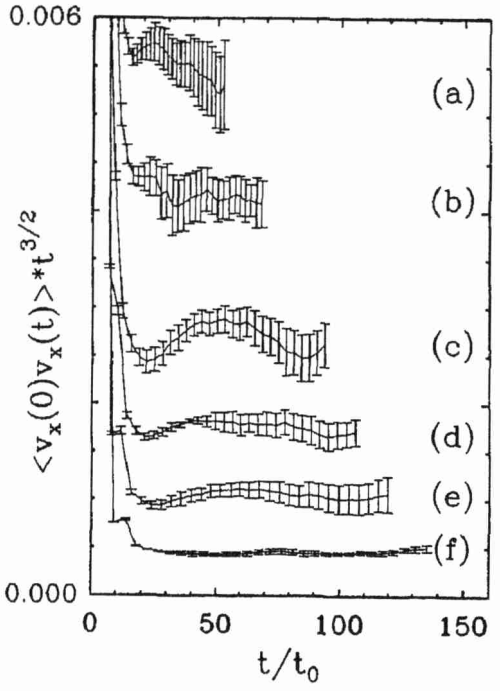


Figure 3: Velocity autocorrelation function multiplied by $t^{3/2}$ for the following densities: (a) $d = 0.2$, (b) $d = 0.3$, (c) $d = 0.5$, (d) $d = 0.6$, (e) $d = 0.7$, (f) $d = 0.8$. The time is in units of mean free time $t_0$.

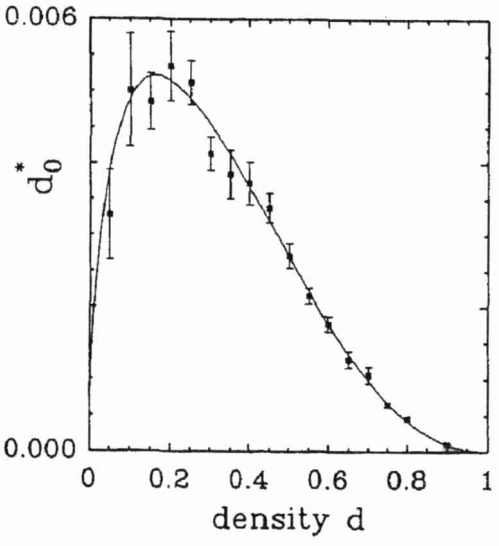


Figure 4: The normalized tail coefficient $d_0^*$ of the tagged-particle VACF for the 3D FCHC system The solid curve is the mode-coupling prediction, the dashed curve is a weighted fit to the simulation data.

the rules are deterministic. Note also that even in 'collisions' that have the same input and output states the tagged particle may still 'collide', i.e. it may change its velocity state.

With these rules, it is obvious that the average velocity of a tagged particle after a collision at site $\mathbf{X}$, depends only on the (colorless) state at that site. In particular, it depends in no way on where the tagged particle was coming from. We can thus define for every (non-empty) site of the lattice the average post collisional velocity that a tagged particle at that site would have at that time:

$$\mathbf{v}_{\mathbf{X}}(t) = \frac{1}{N_{occ}(\mathbf{X})} \sum_{\alpha=1}^{N_{occ}(\mathbf{X})} \mathbf{c}_\alpha \tag{5}$$

where $N_{occ}(\mathbf{X})$ is the total number of particles at site $\mathbf{X}$ and $\mathbf{c}_\alpha$ are the velocities corresponding to the occupied links. At first sight, eqn. 5 may look similar to the expression for the site velocity in a Lorentz gas. The important difference is that for a Lorentz gas the site velocity is fixed by the (time-independent) distribution of random bonds. In contrast, for the LGCA model the site-velocity for a tagged particle changes with every time step. However, apart from this modification, we can use basically the same techniques that worked for the Lorentz gases, to compute the velocity ACF of a tagged particle in a lattice gas cellular automaton.

Consider a tagged particle that is moving at site $\mathbf{X}$ at time $t = 0$, moving with a velocity $\mathbf{c}_\alpha$. At time $t = 1$, the tagged particle will have collided at site $\mathbf{X} + \mathbf{c}_\alpha$ and its average post collisional velocity will be $\mathbf{v}_{\mathbf{X}+\mathbf{c}_\alpha}(t = 1)$. Clearly,

$$< \mathbf{v}(0).\mathbf{v}(1) >= \frac{1}{N_{tot}} \sum_{\alpha,\mathbf{X}} s_\alpha(\mathbf{X}, t = 0)\mathbf{c}_\alpha.\mathbf{v}_{\mathbf{X}+\mathbf{c}_\alpha}(t = 1) \tag{6}$$

where $s_\alpha(\mathbf{X}, t = 0) = 1(0)$ if link $\alpha$ at site $\mathbf{X}$ is occupied (empty) at $t = 0$.

To compute $< \mathbf{v}(0).\mathbf{v}(2) >$, we simply propagate the tagged particle that is at site $\mathbf{X} + \mathbf{c}_\alpha$ to all sites that will be reached from this site in the next time step. For all these sites (denoted by $\{\mathbf{X}(t = 2)\}$ we compute the average post collisional velocity, average it over the set $\{\mathbf{X}(t = 2)\}$ and this yields the average velocity of the tagged particle after two time-steps.

For the actual implementation of this technique the recipe is then the following:

1. compute the average post collisional velocity of the tagged particle at site $\mathbf{X}$, at time $-t$, for all possible $\mathbf{X}$ on the lattice;
2. propagate this average to all sites that will be reached in one cycle from site $\mathbf{X}$;
3. for every site thus reached, compute the average of all the averages that have been propagated to it.
4. iterate steps 2 and 3 until $t$ iterations have been made in total.

Note that in order to compute the velocity correlation function of a tagged particle, use was made of all possible starting positions and trajectories that such a particle could have, compatible with the (deterministic) dynamics of the underlying 'uncolored' lattice gas. The only additional averaging is over all possible time origins and over independent initial conditions. The latter averaging would not be necessary if the lattice gas cellular automaton were strictly ergodic. But as the models studied in this paper are known to have spurious invariants [27], averaging over a number of independent initial conditions is advisable.

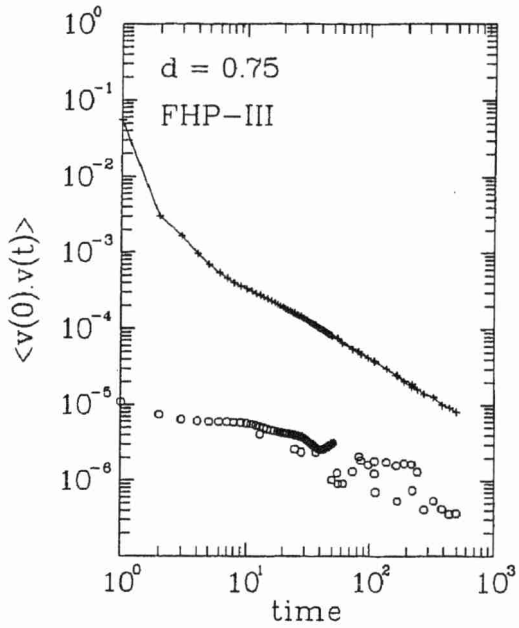


Figure 5: Example of the normalized velocity autocorrelation function of a tagged particle in the in a 2D lattice-gas cellular automaton (FHP-III) at a density $d$=0.75. Note that after an initial rapid decay (and overshoot), the velocity ACF approaches a power-law decay with an exponent $-1$. The estimated error (open circles) decreases with increasing $t$ to a value of order $10^{-6}$. In ref. [12] the statistical error in the long-time tail was of order $0.310^{-2}$.

## 2.5 Models

Simulations of tagged particle diffusion in two-dimensional fluids were carried out for a lattice gas model ('FHP-III', 6 speed-1 particles, one rest-particle, defined in ref. [21]). System sizes of up to $500 \times 500$ lattice points were studied at densities varying from 5% to 75% occupancy. The simulations were either performed on a CYBER 205 vector-computer or, for the larger systems, on a NEC-SX2 super-computer. As a model for $3D$ fluids we used, the face-centered hyper-cubic(FCHC) lattice gas model [18,22]. Whereas FHP-III is discussed extensively elsewhere [18,21] we should explain the exact version of the 3D FCHC-model that we used in our simulations. In this lattice there are 24 possible velocities, so a collision would require a $2^{24}$-word lookup table, which requires a very large shared memory [19]. In the algorithm used in the present paper the 24-bit state is split into two 12-bit sub-states [20], which requires only a small 12-bit lookup table. This splitting can be done in 6 different ways, one of which is choosen randomly at every collision.

The $3D$ simulations were carried out on systems of up to $60 \times 60 \times 60$ lattice points. In all cases correlations were only computed for time intervals less then the shortest time in which any particle could cross the periodic box. This is in contrast to corresponding simulations of long-time tails in atomic fluids [8] where time intervals up to 5 times the acoustic wave traversal time had to be used. In the present simulation the VACF is calculated for different densities varying from $d = 0.05$ to $d = 0.90$, where $d$ is defined as the average number of particles per link per node ($d = \rho/b$). In order to estimate the statistical error of the VACF 5 to 10 independent simulations per density were performed.

## 2.6 Results

### 2.6.1 Three dimensional model

Let us first look at the results for one density $d = 0.1$. In figure 2 we show the VACF for this density. For shorter times we see that the decay is approximately exponential, and for longer times we clearly observe the algebraic decay, which appears in the $\log - \log$ plot as a straight line. A convenient way to present the results for a range of densities is to multiply the VACF with $t^{-3/2}$, see figure 3. If the decay is algebraic with the predicted exponent $-3/2$ these functions should reach a constant value in the limit $t >> t_0$. This behavior is indeed observed. These plateau values should equal the amplitude of the algebraic tail and can be compared with the prediction of mode-coupling theory, see figure 4. This figure shows that there is essentially quantitative agreement between the simulated and the predicted amplitudes for all densities. We wish to stress that there are *no* adjustable parameters in the comparison of theory and simulation.

### 2.6.2 Two dimensional model

Simulations of tagged particle diffusion were carried out for a $2D$ lattice gas model ('FHP-III', 6 speed-1 particles, one rest-particle, with alternating collision rules for odd and even timesteps, see ref. [21]). System sizes of up to $500 \times 500$ lattice points were studied at densities $d$ ranging from 0.05 to 0.75 (i.e. 5% to 75% occupancy per velocity state).

Figure 5 shows the velocity autocorrelation function of a tagged particle in the $2D$ lattice gas at a density $d = 0.75$. The velocity ACF has been normalized to 1 at $t = 0$. Here, and in all other cases shown, correlations were only computed for time intervals less than the shortest time in which any particle could cross the periodic box. In many of

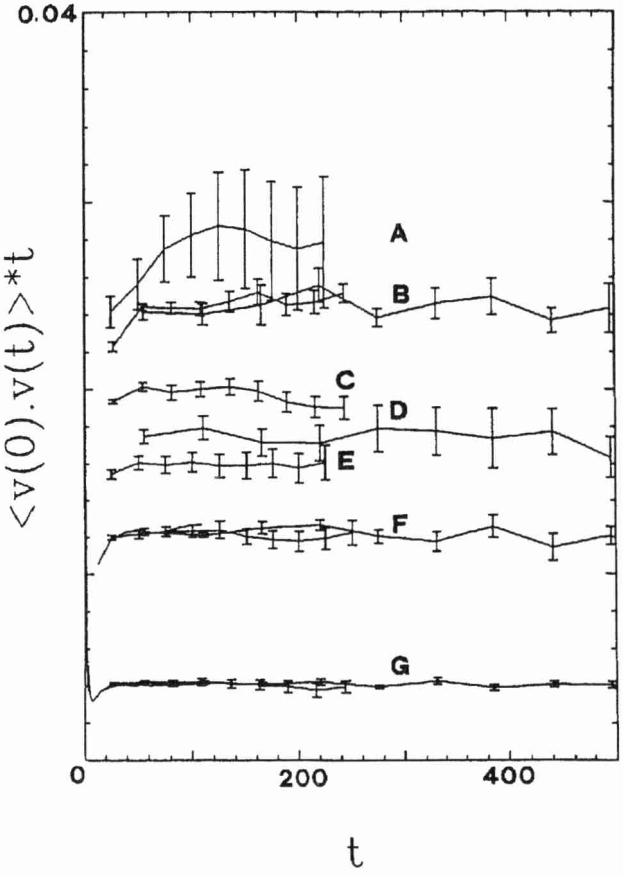


Figure 6: In order to detect possible deviations from the $t^{-1}$ decay in the velocity ACF of a tagged particle in a $2D$ LGCA at long times, this figure shows $t < \mathbf{v}(0).\mathbf{v}(t) > / < v^2 >$ as a function of time $t$. The letters refer to the density: $A - G$ correspond to $d = 0.1, 0.2, 0.3, 0.35, 0.4, 0.5$ and $0.75$ For some densities more than one simulation result is plotted. Within the statistical accuracy of the present calculations (the error bars have a length of 2 standard deviations) no systematic deviations from the $t^{-1}$ decay can be detected. However, our error estimate is probably too conservative because a fit to the simulation data shows that for all 7 points with $d \geq 0.3$ the VACF decays with an effective exponent $\beta > 1$ (up to 3%).

our calculations the statistical error is of order $10^{-6}$, which is about a factor $10^3$ lower than has been achieved with conventional techniques. Note that such an error reduction corresponds to a gain of $10^6$ in computer time. Initially the decay of the velocity ACF is approximately exponential. The characteristic decay time $t_0$ of this exponential ranges from 5.6 at $d = 0.05$ to 0.33 at $d = 0.75$. There is a surprisingly large time-interval where the decay is no longer exponential but not yet algebraic. However, after some 30 collision times the decay appears to become algebraic. In order to see this latter effect more clearly, fig. 6 shows the function $t < \mathbf{v}(0).\mathbf{v}(t) >$ for densities $0.1 \leq d \leq 0.75$. If the velocity ACF decays as $t^{-1}$ then the curves in fig. 6 should approach a constant value as $t \to \infty$. Such behavior is indeed observed. This in itself is maybe not surprising, but it is reassuring as it has been argued that the hydrodynamic long-time tails observed in computer simulation on continuous systems may be due to a propagation of numerical errors [28]. In the present simulation the discrete dynamics of the lattice gas is solved exactly, hence propagation of numerical errors is ruled out as a factor affecting either the power-law tails or, for that matter, any corrections to the latter.

We have compared the measured amplitude of the $t^{-1}$ tail with the predictions of mode-coupling theory [23] adapted to the LGCA. As it turns out, the expression for the amplitude of the $t^{-1}$-tail in a $2D$ LGCA is equal to eqn. 1 multiplied by a factor $(1 - d)$, where the density $\rho$ for continuous fluids must be interpreted as $7d/v_0$, the number density per unit area for the FHP-III model (7 velocity states per site, volume of the unit cell of the triangular lattice $v_0 = \sqrt{3}/2$). The factor $(1 - d)$ is a consequence of the Fermi statistics and guarantees that the state occupied by the tagged particle contains no fluid particle. It should be stressed that the applicability of eqn. 1 to the FHP model is not self-evident because in the FHP-model there exist unphysical hydrodynamic modes (associated with the staggered momentum density, see ref. [27]). These modes can couple to the microscopic stress tensor, thereby affecting the amplitude of the long-time tail of the stress-stress ACF. However, to leading order in $1/t$ the same staggered modes do *not* couple to the tagged particle current.

In figure 7 we compare the simulation results for the amplitude of the $d_0$ with the predictions of mode coupling theory. As can be seen from the figure, the mode coupling predictions are very close to the simulation results. The remaining discrepancy of a few percent is comparable to that found by Kadanoff et al. [27].

Next, consider the behavior of $< \mathbf{v}(0).\mathbf{v}(t) >$ for $t/t_0 \gg 1$. Velocity correlations were studied for times up to $t = 500$, which corresponds to values of $t/t_0$ ranging from $10^2$ to over $10^3$. In this time regime, which has never before been studied numerically, we would expect to observe a $\log(t)/t$ correction to the $1/t$ tail similar to the one predicted by de Schepper and Ernst for the hard-disk model [9]. In continuous fluids, the relative importance of these logarithmic corrections grows linearly with the gas density. If we would assume that the expression for $d_1$ (see eqn. 2) given in ref. [9] also applies in the case of a lattice gas, then we find the following density dependence of the ratio $d_1/d_0$:

$$d_1/d_0 = -d_0\{(\nu_0 + D_0)^{-1} + (4\nu_0)^{-1} + (8(\nu_0 + \zeta_0))^{-1}\} = -0.247d\,, \qquad (7)$$

where $\zeta_0$ is the bulk viscosity and $\rho\zeta_0 = 1/14$ for $\rho = 0$ [21]. A ratio $d_1/d_0$ of this magnitude should be easily observable in the present simulations. However, as can be seen from figure 6 there is no clear evidence for faster than $t^{-1}$ decay at any density. In fact, a $\log - \log$ fit to the long-time tail of the velocity ACF allows us to estimate the ratio $d_1/d_0$ as a function of density. If we assume a linear density dependence of $d_1/d_0$ we can put the

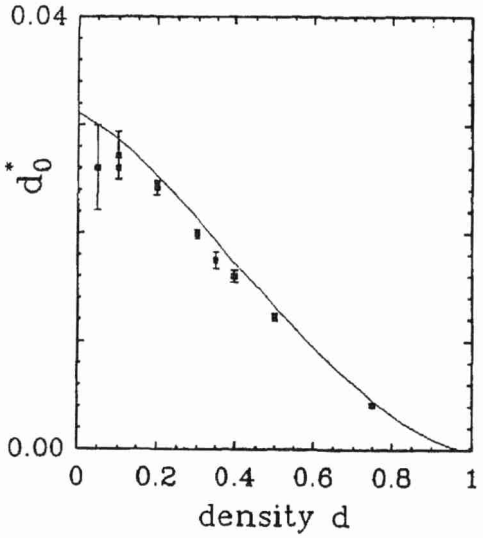


Figure 7: Density dependence of the amplitude of the $t^{-1}$ algebraic tail of the tagged-particle VACF for the two-dimensional FHP-III model. Points: computer simulation results. Drawn curve: mode-coupling theory.

following bounds on the amplitude of this term:

$$d_1/d_0 = -0.02(3)d\,, \tag{8}$$

The kinetic theory estimate for continuous fluids exceeds this value by 7 standard deviations. This suggests that the expression for $d_1/d_0$ given in ref. [9] does not apply to lattice gases. It seems unlikely that this discrepancy is due to the fact that the simulations do not extend to long enough times. More likely, eqn. 7 is not correct for lattice gases, because the the density effects of the Fermi statistics have not been accounted for and the staggered momentum modes that do not contribute to $d_0$ are expected to affect $d_1$. It should be added that even though we compute $< \mathbf{v}(0).\mathbf{v}(t) >$ for $10^2 - 10^3$ mean-free times, we are still well removed from the asymptotic regime where the correlation functions is expected to decay as $1/(t\sqrt{\log t})$.

At present, the lattice-gas equivalent of the prediction for $d_1/d_0$ given in ref. [9] is still lacking. Clearly, such a theoretical result is highly desirable, as it would allow us to decide whether the suppression of significant corrections to the $t^{-1}$ tail is peculiar to lattice gases or if it is indicative of the behavior of $2D$ fluids in general.

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## References

[1] D.Frenkel in: **Liquids, Freezing and the Glass Transition**, J.P.Hansen and D.Levesque (editors), North-Holland, Amsterdam, 1990.

[2] B. J. Alder and T. E. Wainwright, Phys. Rev. **A1**:18(1970).

[3] J.R. Dorfman and E.G.D. Cohen, Phys. Rev. Lett. **25**:1257(1970), Phys. Rev. **A6**:776(1972),**A12**:292(1975).

[4] M.H. Ernst, E.H. Hauge and J.M.J. van Leeuwen, Phys. Rev. **A4**:2055(1971).

[5] Y. Pomeau and P. Résibois, Physics Reports **19**:63(1975).

[6] D. Levesque and W.T. Ashurst, Phys. Rev. Lett. **33**:277(1974).

[7] J.J. Erpenbeck and W.W. Wood, Phys. Rev. **A26**:1648(1982).

[8] J.J. Erpenbeck and W.W. Wood, Phys. Rev. **A32**:412(1985).

[9] I. de Schepper and M. H. Ernst, Physica **87A**:35(1977)

[10] D. Forster, D. R. Nelson and M. J. Stephen, Phys. Rev. **A16**:732(1977).

[11] U. Frisch, B. Hasslacher and Y. Pomeau, Phys. Rev. Lett. **56**:1505(1986).

[12] J.-P. Boon and A. Noullez in: Proceedings of workshop on **Discrete kinetic theory, lattice gas dynamics and foundations of hydrodynamics**, World Scientific, Singapore, 1989.

[13] P.M. Binder and D. d'Humières, Los Alamos preprint LA-UR-1341(1988) and in ref. [24].

[14] J. -P. Boon and A .Noullez, to be published.

[15] D.Frenkel in: Proceedings of workshop on **Cellular Automata and Modelling of Complex Physical Systems**, P. Manneville(editor), Springer, Berlin, 1989.

[16] D.Frenkel and M.H.Ernst, Phys. Rev. Lett. **63**:2165(1989).

[17] D. Frenkel, Phys. Lett. **A121**,385(1987)

[18] U. Frisch, D. d'Humières, B. Hasslacher, P. Lallemand, Y. Pomeau and J.-P. Rivet, Complex Systems **1**:649(1987).

[19] J.-P. Rivet, M. Hénon, U. Frisch and D. d'Humières, Europhys. Lett. **7**:231(1988).

[20] A.J.C. Ladd and D. Frenkel in: Proceedings of workshop on **Cellular Automata and Modelling of Complex Physical Systems**, P. Manneville(editor), Springer, Berlin, 1989.

[21] D. d'Humières and P. Lallemand, Complex Systems **1**:599(1987).

[22] D. d'Humières, P. Lallemand and U.Frisch, Europhys. Lett. **2**:291(1986).

[23] M.H. Ernst in: Proceedings of Les Houches Summerschool on **Liquids, Freezing and the Glass Transition**, J.P.Hansen and D.Levesque (editors), North-Holland, Amsterdam (1990).

[24] Proceedings of workshop on **Cellular Automata and Modelling of Complex Physical Systems**, P. Manneville(editor), Springer, Berlin, 1989.

[25] R. B. Pandey, D. Stauffer, A. Margolina and J. G. Zabolitzky, J. Stat. Phys. **34**:427(1984).

[26] M. E. Colvin, A. J. C. Ladd and B. J. Alder, Phys. Rev. Lett. **61**:381(1988)

[27] L. P. Kadanoff, G. R. McNamara and G. Zanetti, Phys. Rev. **A40**:4527(1989)

[28] R. F. Fox, Phys. Rev. **A27**,3216(1983).